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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of:

Shigetoshi SASANO et al.

Serial No. 09/626,913

Group Art Unit: 1711

Filed: July 27, 2000

Examiner: Ms. Melanie D. Bagwell

For: Laminate Adhesive

DECLARATION UNDER RULE 132

Honorable Commissioner of Patent and Trademarks, Washington, D.C.

Sir:

I, Shigetoshi SASANO declare:

That I am a citizen of Japan, residing at 1-20, Rikyumaemachi 1-chome, Suma-ku, Kobe, Japan;

That I was born on September 14, 1964 in Shiga and graduated from the Department of Applied Chemistry, Faculty of Engineering, Grad-school of Kobe University, Japan in March 1989;

That I have been employed by Takeda Chemical Industries, Ltd., Japan, since April 1989, and engaged in the research in the field of Polyurethane Adhesives;

That I am the first named inventor of U.S. Patent Application Serial No. 09/626,913; and

That the following experiments were conducted by myself according to the Example 1 set forth in the specification of EP0643734B1;

EXPERIMENTS

1. Preparation of Aqueous Polyurethane Polymer Dispersion

A polyurethane prepolymer was first prepared using the following recipe:

	<u>grams</u>	<u>% of Prepolymer Reactants</u>
DESMODUR W*	414.00	46.00
2,2-dimethylol propionic acid	50.40	5.60
PRIPLAST 3192**	394.74	44.19
1,4-cyclohexane dimethanol	37.86	4.21
N-methyl pyrrolidone	300.00	
stannous octoate (catalyst)	0.36	

* 4,4'-dicyclohexylmethane diisocyanate (commercially available).

** a commercially available polyester polyol from Unichema based on a C-36 dimer acid and 1,6-hexanediol.

The above-mentioned ingredients (apart from the stannous octoate) were charged to a reactor and heated, under dry nitrogen, to 50 °C. Half of the stannous octoate was added. The reaction mixture was heated to 80-85°C and kept at this temperature for 30 minutes. The remaining half of the stannous octoate was added to the reactor and the reaction mixture was held at 80-85°C for a further 60 minutes. The residual NCO group content was found to be 4.72%.

The reaction mixture was cooled down to 65 °C and 32.58g triethylamine (TEA) was added to neutralise the carboxyl groups, to thereby produce the polyurethane prepolymer.

After 100g of the resulting prepolymer was dispersed into 81.1g of the ion-exchange distilled water, 1.75g of hydrazine was added to the dispersion to effect chain-extension, to thereby produce the aqueous polyurethane polymer dispersion.

2. Evaluation

The aqueous polyurethane polymer dispersion thus produced was diluted with methanol to prepare a solid content for the use as a laminate adhesive. Using the prepared laminate adhesive, a composite film was produced in a method as mentioned below. The elution test was conducted on the composite film for evaluation.

3. Production of Composite Film

A three-layered composite film of a polyethylene terephthalate film ($12\ \mu\text{m}$ in thickness)/an aluminum foil ($9\ \mu\text{m}$ in thickness)/an unextended polypropylene film ($70\ \mu\text{m}$ in thickness, as subjected to corona discharge treatment) was produced by the following method.

The laminate adhesive was applied onto an aluminum surface of the two-layered composite film made by the polyethylene terephthalate film and the aluminum foil being adhesive bonded together in advance. The spread of the adhesive was $2.5\text{g}/\text{m}^2$ by weight of a solid content of the adhesive per unit area. Then, the surface applied with the adhesive was bonded with the unextended polypropylene film. Thereafter, the bonded film was cured at 50°C for 3 days, for the curing of the adhesive.

4. Elution Test

A bag was made from the composite film thus produced and then was filled with ion-exchange distilled water as the content by the amount of $0.5\text{mL}/\text{cm}^2$ per unit area of the interior surface of the bag. After the bag was sterilized by heated water under the pressure of $19.6 \times 10^4\text{Pa}$ at 120°C for 30 minutes, the contained water was extracted in the solid phase modified with an octadecyl group. Then, a sample was drawn from the extracts which were dissolved in methanol of one hundredth of the quantity of original water. The sample was measured by the gas chromatography (flame ionization detector) and observed on the presence of the cyclic ester compounds and cyclic urethane compounds.

When the eluting substance was observed, the structure of the eluting substance was analyzed by use of a gas chromatograph mass spectrograph. The detection limit of the gas chromatograph (flame ionization detector) for

the methanol solution of dibutyl phthalate was 50 ppb which was equivalent to 0.5 ppb in terms of the concentration of the cyclic ester compounds and cyclic urethane compounds in the extracted water in the elution test.

5. Results

The presence of a substance eluting in the sample was observed by the measurement with the gas chromatograph (flame ionization detector). The structure of the eluting substance was analyzed with the gas chromatograph mass spectrograph, and, as a result, the substance was identified as the cyclic ester compound of adipic acid and 1,6-hexandiol produced by the reaction of adipic acid and 1,6-hexandiol.

I, the undersigned, declare that all statements made herein on my knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, of both, under Section 1001 of Title 18 of the United States Code and such willful false statements may jeopardize the validity of the application or any issuing thereon.

Signed this 21st day of March, 2001.

Shigetoshi Sasano

Shigetoshi SASANO